Advanced Fuel Cell Development

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Objectives

- Develop and validate advanced models for fuel cell analysis and design.
- Apply models for advanced fuel cell development.
- Transfer modeling capability to SECA development teams and Core Technology Program participants.
- Analyze performance and durability of SOFC fuel cells under dynamic loading.
- Evaluate low-cost coating techniques for fuel cell interconnects.
- Perform testing and evaluation of SOFC technologies for SECA.

Key Milestones

- Obtain validation data and compare to model results (2003-2004).
- Develop transient capability in the 3-D modeling tool (2003).
- Collaborate with Georgia Tech Core Technology Program Team to provide detailed SOFC modeling capabilities for use in Georgia Tech SOFC failure model (2003).
- Collaborate with Siemens-Westinghouse Development Program in SOFC model validation and application by comparing model predictions with data from Siemens-Westinghouse for tubular geometry (2003-2005).
- Obtain data on the performance of fuel cells under dynamic loadings and assess the conditions within the cell via transient models (2003-2004).
- Develop dip-coating technique and acquire test data describing performance of perovskite coatings on metallic interconnects (2003).
- Complete the 2003 Future Energy Challenge Inverter Competition (2003).
- Design test facility for evaluating SECA developer prototype units (2003).

Approach

Modeling: NETL has been developing fuel cell models using the FLUENT (Fluent Inc., Lebanon, NH) commercial software code. In this second phase (FY03), improvement was made in the numerical approach to couple electrochemistry and electrical potential field models. This improvement has provided a more computationally efficient tool (i.e. stability is enhanced and solution times have been greatly reduced as a result). The second development phase has also produced a SOFC stack model, allowing for detailed, fully-coupled flow and electrochemical performance predictions for SOFC stacks.

This tool has been made available to SECA developers and to participants in the SECA Core Technology Program [3]. NETL is working with several developers to provide advanced design capability. One key capability of this code is its direct-coupled treatment of electron transport throughout the fuel cell's electrically conductive regions. This capability frees the designer from having to specify/describe arbitrary electron flow through complex geometries (e.g., specially designed flow plates, etc). This is a critical capability to accurately model performance of cells with complex geometries, such as tubular or tube-like cell geometries. Further enhancements to the code during FY 2004 will allow for improved speed and accuracy in the analysis of cells and stacks by enabling the code to run in parallel mode on clusters of computers. In addition, internal reforming modeling capability will be added.

The need for model validation data has been made clear in each of the past SECA workshops. To provide the data needed for validation, NETL has put their SOFC test facility into service in FY 2003. This facility has been designed to test 'generic' fuel cell specimens in partnership with external developers of cells and stacks. Specifically, NETL has collaborated with Dr. Anil Virkar of the University of Utah to acquire specimens tailored to provide known variations in cell properties. The results from testing these cells are now being compared to predictions from our advanced models [4]. This comparison is providing the needed validation of NETL's advanced modeling tools (including the 3-D tool and other reduced-order tools). This data is also available to other developers for use in validating their models.

Dynamic Load Studies: These same tools (both software and experimental) will be used to investigate the performance of fuel cells under dynamic loads during FY 2004-2005. Jorgensen et al. (2000), have shown that chemical kinetics at the cathode can degrade fuel cell performance [2]. While much about basic kinetics at the electrode/electrolyte interface is unknown, it can be expected that these are non-linear, thereby making the steady state results from Jorgensen et al. incomplete when considering actual dynamic loads common in commercial applications (inverter driven or user-load driven). This project will examine these effects and compare them to steady state results to quantify any variances, and to investigate cell properties that may change specifically resulting from dynamic loads.

Interconnect Coatings: Developing a low-cost method to apply a coating to a stainless steel interconnect may enable the use of low cost metalic interconnects for solid oxide fuel cells. This work will experimentally evaluate methods to apply $LaCrO_3$ based coatings for application to SOFC systems. Thin films were deposited by RF-magnetron sputtering on the Cr containing stainless steel (SS) substrates (SS 446). Stainless steel substrates (10x10x5mm) were polished with a diamond spray to a mirror surface. The substrates were coated in the rf sputtering mode under 8x10e-3 Torr Ar+. Highly porous (40 percent) light green color LaCrO3 perovskite was used as a target material. The EDS of an as-deposited film composition gave 56.54/43.46at% La/Cr ratio. After sputtering, the SS sample with deposited film was annealed at various temperatures in air. The heat treated samples were analyzed at each step by XRD, SEM and RAMAN spectroscopy. Additional methods, such as dip-coating, are being compared to evaluate how a successful coating material can be applied in a thin film suitable for the SOFC interconnect.

Test and Evaluation: The Solid State Energy Conversion Alliance (SECA) Program requires the evaluation of prototype fuel cell systems being developed by industry teams. The program has been divided into three phases and following each phase the industry teams will provide the National Energy Technology Laboratory (NETL) with fully functional prototypes for Department of Energy evaluation. A test facility to perform the evaluation has been designed by Concurrent Technologies Corporation and follows industrial test codes for fuel cells. The requirements for the facility were established through communication with the industrial teams who provided overall performance and operational targets for their systems. Construction of the facility will also occur in phases, and is expected to be ready for the first prototype tests in 2005. The testing will independently evaluate how well the industrial teams are meeting their technical targets. The results of the evaluations will be made available to the public through the technical literature.

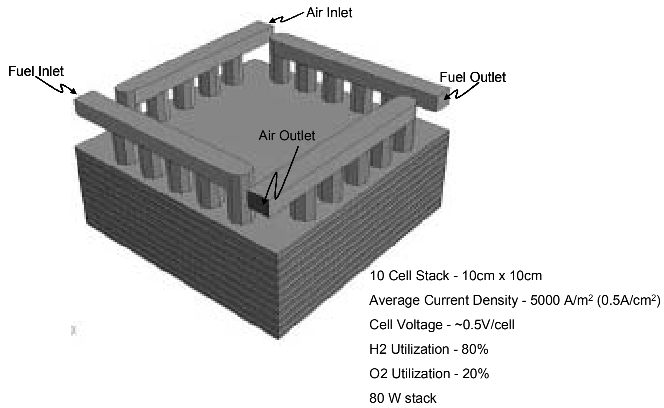

Figure 1. *Prototype SOFC stack.*

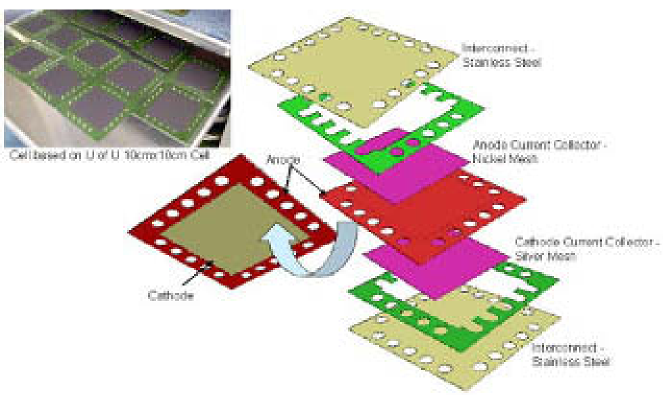

Figure 2*. Detail of stack cell construction.*

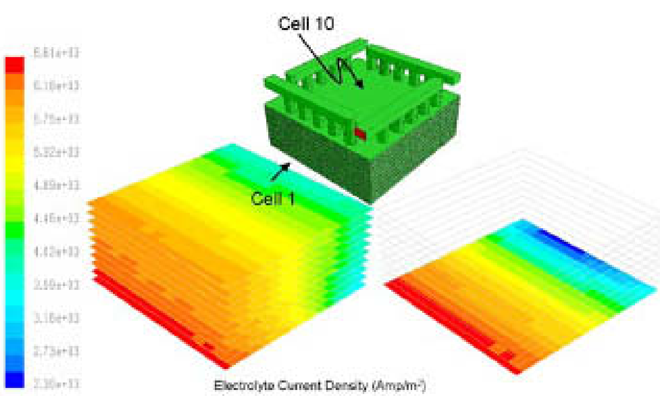

Figure 3. *Contours of predicted current density on selected stack cells.*

Results

Modeling: Results from a detailed modeling study of a prototype short stack of SOFC cells operating in a crossflow configuration are shown in Figures 1-3. These results were used to verify that the stack model produced physically realistic results. Figure 1 shows the basic geometry and model setup used for this analysis. This is a prototype short stack configuration based on a 10 cm x 10 cm cell produced by the University of Utah. Figure 2 shows the details of the individual cell, current collector and seals used in 10-cell stack, and it demonstrates the detailed resolution incorporated in this model. Figure 3 shows selected current density contours at two levels in the stack. Poor distribution of reactants to the individual cells is demonstrated in these results by noting the difference in contours at the two extreme ends of the stack. Validation of stack modeling capabilities will use data from the NETL test facility in 2004.

Validation of the single cell model is being performed in collaboration with researchers at Siemens-Westinghouse Power Corporation (SWPC). The model is being used to simulate SWPC single-cell experiments studying their tubular cell design. Figure 4 shows the geometric details of the single tubular cell being simulated. Figure 5 shows selected views of the current density contours on the electrolyte as a selected operating condition. The large gradient in current density that is observed showcases the code is capable of dealing with the complex cell geometry exhibited by the tubular cell.

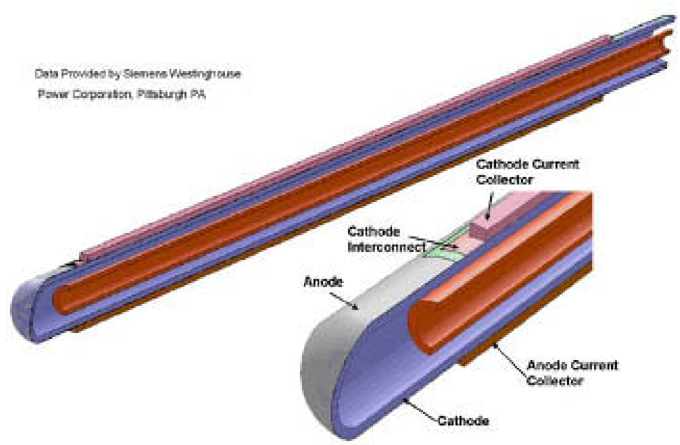

Figure 4. *Detail of Siemens-Westinghouse tubular cell providing data for model validation.*

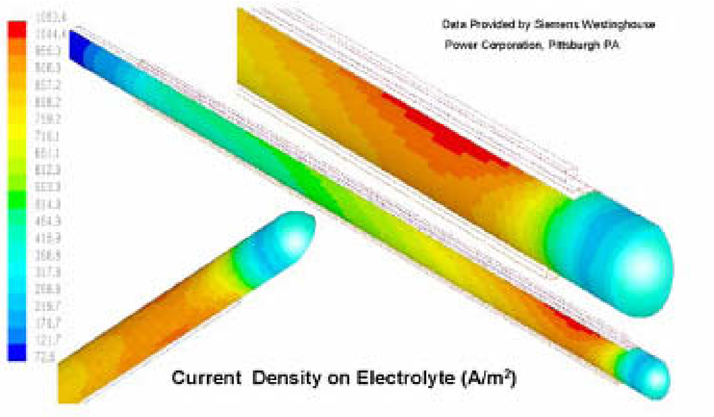

Figure 5. Current Density on Siemens-Westinghouse tubular cell from NETL SOFC model.

Figure 6a. Example of NETL SOFC test stand furnace.

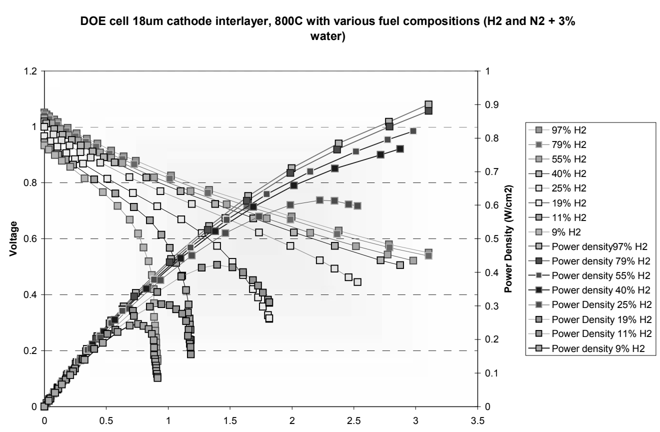

Figure 6b. Data from NETL SOFC test facility illustrating impact of N_2 dilution of fuel.

Model validation is a critical requirement for reliable and accurate use of any model. To achieve these model requirements, NETL is using the new SOFC test facility to acquire experimental data from well-characterized experiments for button cells. Testing for small stacks is planned for 2004. Figure 6a shows a sample cell/stack test rig that will be used for this project. Several other such test rigs will be employed for this project as well. Figures 6b shows performance curves generated in the NETL facility for the standard button-cell geometry with various nitrogen dilution levels for the fuel. This data is being used to validate the detailed NETL model.

Dynamic Load Studies: The effect of inverter ripple current on cell operating conditions was studied [1]. The results are shown in Figures 7 and 8 for 1 mm thick anode supported cell and cathode supported cell, respectively. Figure 7 shows the deviation in hydrogen concentration at the electrode-electrolyte interface (at its extreme) from equivalent steady state cell loading values. As can be seen, for 120 Hz, 0.8 utilization, the deviation in hydrogen concentration increases as current loading or ripple increases. Figure 8 shows the same results, but for oxygen concentration at the cathode. For a fixed 3 percent departure limitation, the ripple must be limited to conditions below the curves shown in Figure 9 for a selected range of current loadings.

Interconnect Coatings: The feasibility of the deposition of La-Cr-O thin film by RF magnetron sputtering was studied. The 40 nm/hour deposition rate was obtained and 5 hours of the deposition was required to grow 0.2mm thin film. The as-deposited film was amorphous as confirmed by XRD. After annealing at 700 °C for 1 hour, the amorphous film transformed to $LaCrO_3$ perovskite structure. Two transformations typically occur during the amorphous to perovskite structure transition: first is the amorphous to $LaCrO_4$ monoclinic structure transformation at 495 °C, and the second is a $LaCrO_4$ to $LaCrO_3$ orthorhombic structure transformation at 780 °C. (However, in our case the temperature of the second transformation was lowered to 700 °C perhaps because of the preheating of the thin film sample at 300 and 500 °C.) The nano-crystalline self-assembled dendritic structure of $LaCrO_3$ perovskite thin film is demonstrated by the SEM shown in Figure 10. Future work in this area will focus on trying to avoid the $LaCrO_4$ phase as an intermediate, and thus obtaining a denser protective coating.

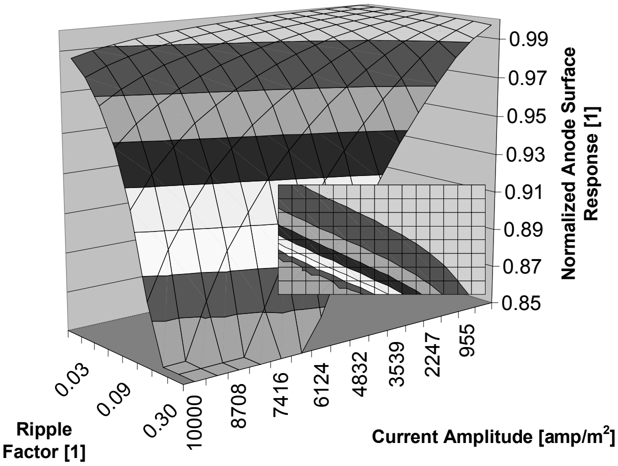

Figure 7. *Hydrogen concentration response for 120 Hz. Average utilization fixed at 0.8.*

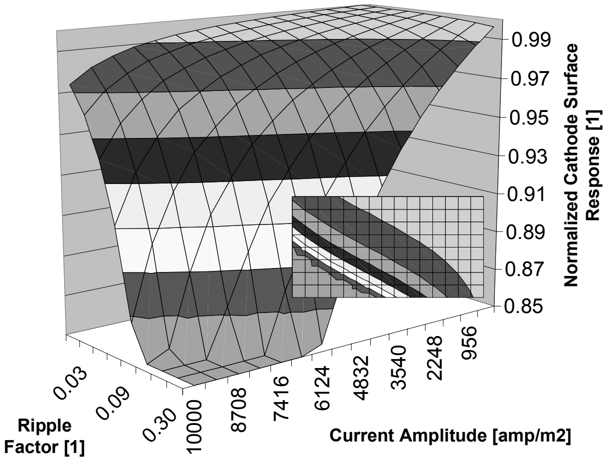

Figure 8. *Oxygen concentration response for 120 Hz. Average utilization fixed at 0.25.*

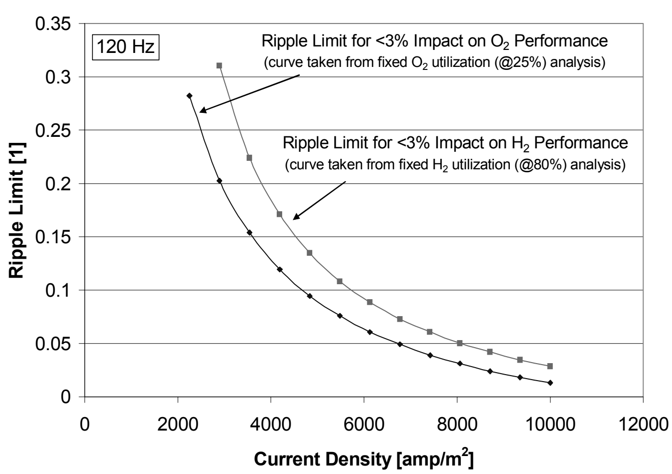

Figure 9. *Limit for ripple for 3% deviation limitation in electrode-electrolyte interface concentration.*

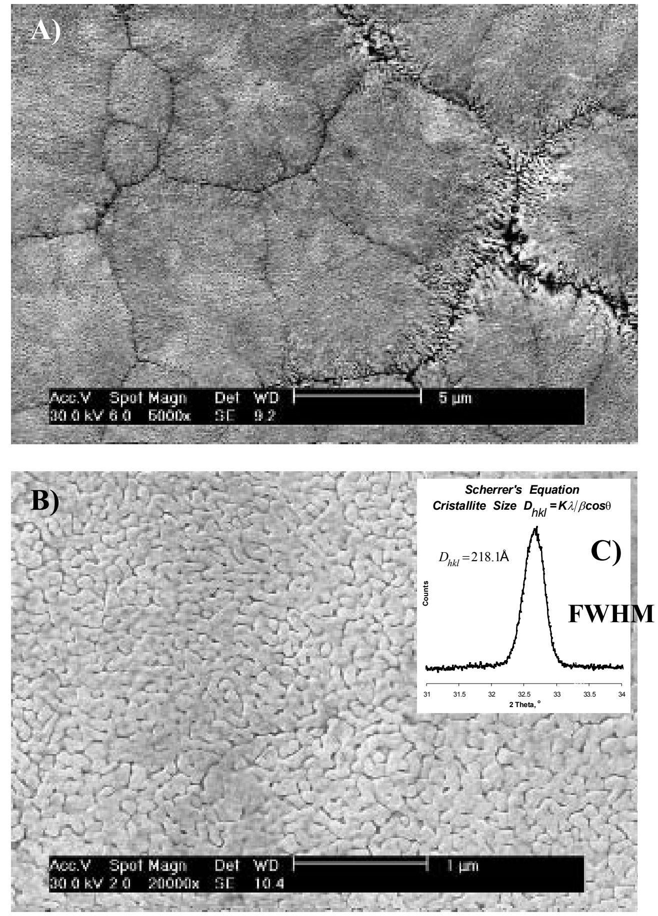

Figure 10. *Micrographs of the $LaCrO_3$ perovskite structure formed after annealing of amorphous film at 700 °C for 1 hour. A) A dendritic structure of $LaCrO_3$ perovskite; B) A self-assembled structure of $LaCrO_3$ crystallines inside grains; C) Inset: Full width at half maximum of {111} peak and calculated crystallite size of the $LaCrO_3$ this film.*

Test and Evaluation: The overall design of the SOFC test facility was completed, shown in Figure 11, and work on Phase I construction was begun. The present facility allows for testing fuel cells using natural gas or propane. Early use of the facility has already been made thorough NETL's support of the 2003 Future Energy Challenge. The goal of the Future Energy Challenge is to develop highly efficient low cost inverter technology suitable for SECA units (>94% efficiency and <$40/kW). In May, NETL tested five inverters, but determined that none of them were presently able to meet these technical targets. Finally, the test facility will also be used in the coming months to perform testing and evaluation of an EPA remote site SOFC power unit being developed by Fuel Cell Technologies, Ltd. During this testing, the performance and application readiness of the product will be evaluated.

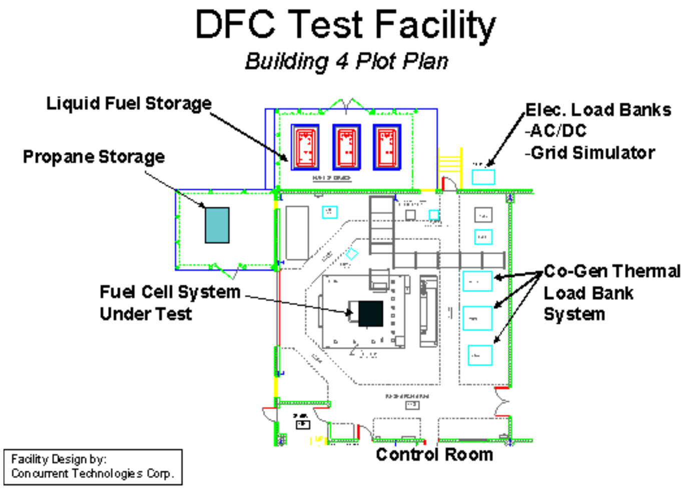

Figure 11. *DOE fuel cell test facility plan. This facility will be used to evaluate SECA prototype units coming from the SECA vertical team developers.*

Conclusions

Great progress is being made to provide new advanced tools for fuel cell design. Basic model development is complete with enhancements underway, providing additional capabilities. These tools are now being validated using data from SOFC developers and from NETL test facilities. Collaboration with SECA developers and Core Technology participants is underway to validate the model and to provide the tools for their use. Validation of the model for single cell and cell stacks will be continued.

Progress is also being made to understand the impact of dynamic loadings on fuel cell operating conditions and materials. Analytic results indicate that significant deviations from steady state conditions are imposed by dynamic loadings such as inverter loads, especially at high average current loadings. As a result, inverter ripple may become a limitation to achieving high current density cells (>1 amp/cm^2) if it is found that the high deviations from steady state loadings cause increased degradation of fuel cell materials. In future work, the test facility will be used to experimentally investigate the effects of dynamic loading of solid oxide fuel cells.

Progress is also being made to understand the ability of using perovskite coatings to achieve low cost interconnects. The present work has shown the basic transformation behavior of LaCrO3 perovskite coatings under conditions typical of SOFC fuel cell operation. This activity provides the ground work for further research that will improve these films through the use of various coating methods, as well as application of dopants (e.g., calcium).

Finally, progress is being made to support evaluation of progress in meeting SECA program objectives. Initial construction and application of a prototype fuel cell test facility is underway, and will be ready by 2005 so that the first (Phase I) SECA prototype units can be evaluated.

References

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